Nondestructive Evaluation of Microstructure of SiC_f/SiC Composites by X-Ray Computed Microtomography

Weon-Ju Kim[†], Daejong Kim, Choong Hwan Jung, Ji Yeon Park, and Lance L. Snead*

Nuclear Materials Division, Korea Atomic Energy Research Institute, Daejeon 305-353, Korea

**Materials Science and Technology Division, Oak Ridge National Laboratory, Oak Ridge, TN 37831, USA*

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ABSTRACT

Continuous fiber-reinforced ceramic matrix composites (CFCCs) have a complex distribution of porosity, consisting of interfiber micro pores and interbundle/interply macro pores. Owing to the complex geometry of the pores and fiber architecture, it is difficult to obtain representative microstructural features throughout the specimen volume with conventional, destructive ceramographic approaches. In this study, we introduce X-ray computed microtomography (X-ray μ CT) to nondestructively analyze the microstructures of disk shaped and tubular SiC_f/SiC composites fabricated by the chemical vapor infiltration (CVI) method. The disk specimen made by stacking plain-woven SiC fabrics exhibited periodic, large fluctuation of porosity in the stacking direction but much less variation of porosity perpendicular to the fabric planes. The X-ray μ CT evaluation of the microstructure was also effectively utilized to improve the fabrication process of the triple-layered tubular SiC composite.

Key words : X-ray computed microtomography, SiC_f/SiC composite, Microstructure, Nondestructive evaluation

1. Introduction

Silicon carbide fiber-reinforced silicon carbide matrix (SiC_f/SiC) composites have been studied extensively for structural materials in various fields such as advanced engines, gas turbines, heat exchangers, and defense industries.¹⁾ SiC_f/SiC composites are also being considered as structural components for advanced nuclear energy systems such as fusion and advanced fission reactors owing to their excellent high-temperature properties, irradiation tolerance, inherent low activation and other superior physical/chemical properties.²⁻⁴⁾ Recently, there have also been efforts in applying SiC_f/SiC composites to fuel claddings of various types of reactors, such as liquid salt-cooled advanced high temperature reactors (LS-AHTRs), gas-cooled fast reactors (GFRs), sodium-cooled fast reactors (SFRs), and light water reactors (LWRs).⁵⁻⁸⁾

In general, the chemical vapor infiltration (CVI) technique has been used most widely to produce SiC_f/SiC composites, especially for use in the in-core components of nuclear reactors.^{1,8)} Although the technique produces a very pure SiC matrix and thus an irradiation tolerant composite, it results in a complex distribution of porosity. The pores consist of interfiber micro pores and interbundle/interply macro pores. The complex geometry of pores and fiber architecture also induces a highly anisotropic behavior of the composite

properties. The inhomogeneity of the fiber-reinforced composites makes it difficult to obtain representative microstructural features throughout the specimen volume with the conventional, destructive ceramographic techniques. Therefore, nondestructive approaches would be better for characterizing and defining the locally inhomogeneous microstructure of SiC_f/SiC composites. Among various non-destructive evaluation techniques, e.g., the ultrasonic method, infrared thermography, X-ray computed microtomography (X-ray μ CT), neutron tomography, and so on,⁹⁻¹²⁾ the X-ray μ CT is quite useful for characterizing the 3-dimensional (3D) microstructure of fiber-reinforced composites at micro and even nano scales^{13,14)} in terms of simplicity, resolution, and affordability.

Through the development of high-resolution X-ray μ CT techniques, X-ray μ CT has recently become a widely used technique for microstructural characterization based on a synchrotron-generated or laboratory micro X-ray source. The X-ray μ CT has been used to investigate the evolution of damage in situ during mechanical tests, such as tensile, creep, and fatigue tests, for graphite, metallic alloys, polymer/metal matrix composites, and other various material systems.^{13,15-19)} The technique has been proven to be a powerful tool for analyzing microstructures of highly porous cellular structures as well.²⁰⁻²²⁾ Different morphological parameters of porous materials, such as density distribution and cell and pore size, are not easy to measure through conventional 2D characterization methods.²⁰⁾ The X-ray μ CT has also been used to explore the microstructures of fiber-reinforced ceramic matrix composites, such as C_f/C, C_f/SiC, and SiC_f/SiC composites.²³⁻²⁸⁾ Kinney et al.²⁵⁾ used the X-ray μ CT to

[†]Corresponding author : Weon-Ju Kim

E-mail : weonjkim@kaeri.re.kr

Tel : +82-42-868-8029 Fax : +82-42-868-8549

measure the growth of a SiC matrix in a woven Nicalon-fiber composite during a CVI process. They were able to measure the degree of densification within individual fiber tows and follow the closure of macroscopic pores during successive CVI steps. Bale et al.²⁸⁾ performed 3D observations of damage evolution in SiC_f/SiC composites using a synchrotron-based X-ray μ CT and resolved sequences of microcrack damage in situ as cracks grew under load at high temperatures of up to 1750°C.

In this study, we used the X-ray μ CT method to nondestructively analyze the microstructure of SiC_f/SiC composites. The pore structure and porosity distribution in different directions were analyzed for a plain-woven disk shaped and a wound tubular SiC_f/SiC composite specimen.

2. Experimental Procedure

Two kinds of SiC_f/SiC composite specimens were used for the X-ray μ CT analyses, disk shaped and triple-layered (triplex) tubular composites; the schematic configurations are shown in Fig. 1. The disk specimen was machined from a 2-dimensional (2D) plain-woven composite plate reinforced with Hi-NicalonTM (Nippon Carbon, Japan) SiC fibers. The matrix was infiltrated by the CVI method and the specimen dimensions were 6 mm in diameter and 2.8 mm in thickness. The tubular specimen consisted of three layers. The innermost layer was a high-density monolithic CVD SiC and the second layer was a SiC fiber-reinforced SiC matrix (SiC_f/SiC) composite to increase the mechanical properties and prevent brittle fracture of the composite tube. We used Tyranno SA3 (Ube Ind., Japan) fibers to reinforce the composite layer. Another CVD SiC was finally coated to protect the relatively porous composite layer containing a pyrolytic carbon (PyC) interphase from corrosion. This three-layer structure of the SiC_f/SiC composite tube is one of the concepts of advanced nuclear fuel claddings with a superior

accident tolerance, being currently of great interest worldwide.^{7,8)} The typical dimensions of the three-layer tube used in this study were 9 mm in the inner diameter with a wall thickness of about 1 mm and a length of 50 mm.

In the X-ray μ CT technique, several hundred or more than a thousand 2D X-ray radiographic images are taken at different angles by rotating the specimen. Then, using 3D reconstruction software, the spatial distribution of the absorption coefficient within the sample is reconstructed to produce a 3D volume image of the specimen. In this study, we used a laboratory-based polychromatic X-ray source, and Al/Cu filters were used to reduce the beam hardening effect of the polychromatic X-ray beam. For the disk sample, the spatial resolution (image pixel size) of the X-ray μ CT equipment (microXCT, Xradia, USA) was 8 μ m at a field of view of 4.5 mm. The typical X-ray power was 100 kV, 80 μ A with an exposure time of 15 seconds per image, and the number of acquired radiographic projections were 361 collected at 0.5° angular steps over a 180° rotation of the specimen. Fig. 2 shows an example of the experimental set up of the X-ray μ CT equipment. In the case of a tubular sample, for which we used different equipment (Skyscan1172, Bruker, Belgium), the spatial resolution was 2.9-13.3 μ m and 181-361 radiographic images were collected at angular steps of 0.5-1.0° depending on the field of view.

3. Results and Discussion

Figure 3 shows the 3D volume image of the disk specimen reconstructed from the radiographic images. Lighter and darker regions of the 3D rendered volume in Fig. 3(a) correspond to the SiC matrix/fiber and pores, respectively. We can also extract the 3D image of pores from the volume images shown in Fig. 3(b). The 3D image of pores was obtained by making the voxels (pixels in 2D images) belonging to the SiC matrix/fiber phases transparent. It can be seen from the 3D pore image that some macro pores are interconnected by channel-shaped pores (the circled regions) and some other

Fig. 1. Schematic configurations of SiC_f/SiC composite samples used in this study, (a) disk shape and (b) triplex tubular composites.

Fig. 2. A photograph showing the experimental set up of X-ray μ CT equipment.

Fig. 3. 3D volume images of (a) the SiC_f/SiC disk specimen reconstructed from the radiographic images and (b) pores extracted from the reconstructed volume of the composite.

Fig. 4. Typical sliced images (a) in the transaxial and coronal directions and (b) the binarized images through the threshold operation of the sliced images.

pores are closed. The macro pores are highly tortuous and rather flattened parallel to the fabric's plane.

From the volume image, we can obtain a number of cross-sectional sliced images along various directions, e.g., transaxial (XY), coronal (YZ), and sagittal (XZ) planes. To evaluate the distribution of porosity in the sliced images, each sliced image was binarized into two phases (pore and solid phases) through a threshold operation using ImageJ software.²⁹⁾ Figure 4 shows typical sliced images in the XY and XZ directions and the binarized images through the threshold operation. The binarization was performed by selecting an appropriate black level from the black and white contrast histogram while setting the white level to 0. Fiber bundles along the warp and weft directions in the woven fabric can be clearly seen from the sliced images. It can also be seen that there are two types of pores, interfiber micro pores and interbundle macro pores. However, it is difficult to clearly observe the pores with submicron or several micron sizes within fiber bundles due to the limitation of resolution used in this experiment. If we use a smaller sample around 1 mm, we can obtain a higher resolution and detect submicron pores. In this study, however, we used a larger sample (6 mm in diameter) at the expense of the resolution because we intended to analyze the overall distribution of porosity of the 2D stacked composite.

Figure 5 shows the porosity distribution of the SiC_f/SiC composite calculated from the binarized images in the three directions, XY, XZ, and YZ slices. In the transaxial direc-

Fig. 5. Porosity distribution of the SiC_f/SiC composite calculated from the sliced images in three directions.

Fig. 6. X-ray μ CT images corresponding to the slice numbers (a) 102, (b) 189 in the XY slices, (c) 175 in the XZ slices, and (d) 172 in the YZ slices in Fig. 5.

tion, it shows a significant variation in the porosity due to the stacking nature of the 2D composite, a lower porosity within the plies and a higher porosity between the plies. During the weaving and stacking of the SiC fiber fabrics, the fiber bundles are gently pressed and thus have a lenticular cross-sectional shape, as shown in the lower image of Fig. 4(a). Therefore, in the transaxial direction, the porosity shows the lowest value approximately at the central plane of a ply and continuously increases upon approaching the interface between plies. The peaks (or valleys) of porosity in the transaxial direction are periodically repeated at about every 25 slices on average. Because the distance between slices is 8 μ m in this experiment, the peak-to-peak distance is calculated to be about 0.2 mm, being close to the thickness of one ply. On the contrary to the transaxial direction, the XZ and YZ slices show less variation in porosity than the transaxial slices and do not exhibit an obviously periodic fluctuation of porosity.

Figures 6(a) and (b) represent the images of slice numbers 102 and 189 in the transaxial direction, corresponding to the lowest and highest porosity planes in Fig. 5, respec-

Fig. 7. 3D reconstructed volume images of the triplex SiC composite tube viewed at different angles. The dot-to-dot distance in the scale marker box corresponds to 1 mm.

Fig. 8. Typical X-ray μ CT sliced images along (a) the transaxial and (b) coronal planes of the triplex SiC composite tube.

tively. In slice 102 close to the central plane of a ply, the fiber bundles are closely packed and the interbundle pore sizes are less than 200 μ m. On the other hand, there are very large interbundle pores with a size of about 1 mm in slice 189 close to the interply region. The images in the XZ and YZ slices (Figs. 6(c) and (d)) show less variation of porosity, as previously mentioned.

We also analyzed the microstructure of a triplex SiC composite tube using the X-ray μ CT. Figure 7 shows the 3D volume of a SiC composite tube viewed at different angles. Typical sliced images along the transaxial and coronal planes are also shown in Fig. 8, which are acquired at a resolution of 3.8 μ m. It can be seen from both figures that the inner CVD SiC tube shows a very uniform thickness throughout the tube length. However, the outer surface of the composite tube depicts a rather rough feature. This is attributed to the fracture of individual fibers during the winding step of the fiber yarn. The brittle nature of the SiC ceramic fiber in spite of the fine diameter (7.5 μ m for Tyranno SA3) makes the fiber susceptible to fracture during the winding process even under a small amount of tension, if the winding process is not properly controlled. The deposition of the SiC matrix on the fractured fibers induces a rough surface as shown in the lower left region of Fig. 8(a). The surface roughness is an important factor for the application of nuclear fuel cladding because it affects the cooling efficiency of water coolant flowing with high velocity.

It can also be seen from Fig. 8 that there are large circum-

Fig. 9. A sliced image in the coronal plane sectioning through the composite layer of the triplex SiC composite tube.

Fig. 10. X-ray μ CT sliced images along (a) the transaxial and (b) coronal planes of the triplex SiC composite tube fabricated by an improvement of the winding process.

ferential voids at the interface between the inner CVD SiC and the composite layers (e.g., the upper right region of Fig. 8(a)) and within the composite layer. If the winding process is not adequately controlled, there can be a large space between fiber bundles unwound with fibers. It is difficult to sufficiently infiltrate these large voids with the SiC matrix during the CVI step. Figure 9 shows a sliced image in the coronal plane sectioning through the composite layer. For the triplex composite tube shown in Figs. 7-9, the fibers were first wound at a 90° angle on the CVD SiC inner layer and another layer was wound over the first layer at a different angle of $\pm 55^\circ$ with respect to the long axis of the tube. As a result, we can observe the fibers wound at different angles, 90° and $\pm 55^\circ$, in Fig. 9. We can also observe large interbundle voids with a size of about 1 mm and resolve some individual fibers owing to the relatively high resolution used in this analysis. From the lessons learned through these analyses, we carefully controlled the fiber winding process to minimize the fiber fracture and reduce the interbundle void space. Figure 10 shows typical sliced images along the transaxial and coronal planes of the triplex composite tube fabricated by an improvement of the winding process. It can be seen that the surface roughness is greatly improved and the size and number of voids are also significantly reduced compared to the composite tube shown in Fig. 8. Detailed analyses on the effect of the fiber winding method on the surface roughness and fiber fraction are given in our separate publication.³⁰⁾

4. Summary and Perspectives

X-ray μ CT is a powerful tool for analyzing materials with

an inhomogeneous structure such as continuous fiber-reinforced ceramic matrix composites. We used the X-ray μ CT method to nondestructively analyze the microstructure of SiC_f/SiC composites with disk and tubular shapes. The technique was able to characterize the distribution of interfiber and interbundle/interply porosities throughout the specimen volume without a time consuming and destructive sample preparation. The X-ray CT data were also used to improve the fabrication process of the triplex SiC composite tube for application in accident-tolerant nuclear fuel claddings. The technique is expected to be used for in-situ mechanical tests or performance tests under severe accident conditions of nuclear reactors to explore the crack initiation and propagation behavior. It is also expected that the digitally rendered data of a 3D volume can be useful to provide a realistic model for simulation studies such as finite element analyses of SiC_f/SiC composites with anisotropic thermal and physical properties.

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REFERENCES

1. R. Naslain, "Design, Preparation and Properties of Non-Oxide CMCs for Application in Engines and Nuclear Reactors: an Overview," *Compos. Sci. Technol.*, **64** [2] 155-70 (2004).
2. L. L. Snead, T. Nozawa, M. Ferraris, Y. Katoh, R. Shnavski, and M. Sawan, "Silicon Carbide Composites as Fusion Power Reactor Structural Materials," *J. Nucl. Mater.*, **417** [1-3] 330-39 (2011).
3. W. -J. Kim, S. M. Kang, K. H. Park, A. Kohyama, W. -S. Ryu, and J. Y. Park, "Fabrication and Ion Irradiation Characteristics of SiC-Based Ceramics for Advanced Nuclear Energy Systems (in Korean)," *J. Kor. Ceram. Soc.*, **42** [8] 575-81 (2005).
4. B. Riccardi, L. Giancarli, A. Hasegawa, Y. Katoh, A. Kohyama, R. H. Jones, and L. L. Snead, "Issues and Advances in SiC_f/SiC Composites Development for Fusion Reactors," *J. Nucl. Mater.*, **329-333** 56-65 (2004).
5. Y. Katoh, L. L. Snead, I. Szlufarska, and W. J. Weber, "Radiation Effects in SiC for Nuclear Structural Applications," *Curr. Opin. Solid State Mater. Sci.*, **16** [3] 143-52 (2012).
6. L. Chaffron and J. -L. Seran, "Innovative SiC_f/SiC Composite Materials for Fast Reactor," *Nuclear Fuels and Structural Materials for the Next Generation Nuclear Reactors*, San Diego, USA, June 13-17, 2010.
7. K. Yueh, D. Carpenter, and H. Feinroth, "Clad in Clay," *Nucl. Eng. Int.*, **55** 14-16 (2010).
8. W. -J. Kim, D. Kim, and J. Y. Park, "Fabrication and Material Issues for the Application of SiC Composites to LWR Fuel Cladding," *Nucl. Eng. Technol.*, **45** [4] 565-72 (2013).
9. D. E. Bray and D. McBride, *Nondestructive Testing Techniques*, John Wiley & Sons, New York, 1992.
10. J. Kim, P. K. Liaw, and H. Wang, "The NDE Analysis of Tension Behavior in Nicalon/SiC Ceramic Matrix Composites," *JOM*, **55** [1] 1-13 (2003).
11. S. R. Stock, "Recent Advances in X-Ray Microtomography Applied to Materials," *Int. Mater. Rev.*, **53** [3] 129-81 (2008).
12. M. Strobl, I. Manke, N. Kardjilov, A. Hilger, M. Dawson, and J. Banhart, "Advances in Neutron Radiography and Tomography," *J. Phys. D: Appl. Phys.*, **42** [24] 243001 (2009).
13. F. Beckmann, R. Grupp, A. Haibel, M. Huppmann, M. Nothe, A. Pyzalla, W. Reimers, A. Schreyer, and R. Zettler, "In-Situ Synchrotron X-Ray Microtomography Studies of Microstructure and Damage Evolution in Engineering Materials," *Adv. Eng. Mater.*, **9** [11] 939-50 (2007).
14. D. Attwood, "Nanotomography Comes of Age," *Nature*, **442** [7103] 642-43 (2006).
15. J. Baruchel, J. -Y. Buffiere, P. Cloetens, M. D. Michiel, E. Ferrie, W. Ludwig, E. Maire, and L. Salvo, "Advances in Synchrotron Radiation Microtomography," *Scr. Mater.*, **55** [1] 41-46 (2006).
16. A. Hodgkins, T. J. Marrow, P. Mummery, B. Marsden, and A. Fok, "X-Ray Tomography Observation of Crack Propagation in Nuclear Graphite," *Mater. Sci. Technol.*, **22** [9] 1045-51 (2006).
17. Y. -C. Hung, J. A. Bennett, F. A. Garcia-Pastor, M. Di Michiel, J. -Y. Buffiere, T. J. A. Doel, P. Bowen, and P. J. Withers, "Fatigue Crack Growth and Load Redistribution in Ti/SiC Composites Observed In Situ," *Acta Mater.*, **57** [2] 590-99 (2009).
18. H. Zhang, H. Toda, P. C. Qu, Y. Sakaguchi, M. Kobayashi, K. Uesugi, and Y. Suzuki, "Three-Dimensional Fatigue Crack Growth Behavior in an Aluminum Alloy Investigated with In Situ High-Resolution Synchrotron X-Ray Microtomography," *Acta Mater.*, **57** [11] 3287-300 (2009).
19. P. J. Schilling, B. R. Karedla, A. K. Tatiparthi, M. A. Verges, and P. D. Herrington, "X-Ray Computed Microtomography of Internal Damage in Fiber Reinforced Polymer Matrix Composites," *Compos. Sci. Technol.*, **65** [14] 2071-78 (2005).
20. C. Petit, S. Meille, and E. Maire, "Cellular Solids Studied by X-Ray Tomography and Finite Element Modeling - a Review," *J. Mater. Res.*, **28** [17] 2191-201 (2013).
21. R. C. Atwood, J. R. Jones, P. D. Lee, and L. L. Hench, "Analysis of Pore Interconnectivity in Bioactive Glass Foams Using X-Ray Microtomography," *Scr. Mater.*, **51** [11] 1029-33 (2004).
22. E. Maire, P. Colombo, J. Adrien, L. Babout, and L. Biasetto, "Characterization of the Morphology of Cellular Ceramics by 3D Image Processing of X-Ray Tomography," *J. Eur. Ceram. Soc.*, **27** [4] 1973-81 (2007).
23. J. Martin-Herrero and C. Germain, "Microstructure Reconstruction of Fibrous C/C Composites from X-Ray Microtomography," *Carbon*, **45** [6] 1242-53 (2007).
24. J. M. Hausherr, W. Krenkel, F. Fischer, and V. Altstadt, "Nondestructive Characterization of High-Performance C/

- SiC-Ceramics Using X-Ray-Computed Tomography," *Int. J. Appl. Ceram. Technol.*, **7** [3] 361-68 (2010).
25. J. H. Kinney, T. M. Breunig, T. L. Starr, D. Haupt, M. C. Nichols, S. R. Stock, M. D. Butts, and R. A. Saroyan, "X-Ray Tomographic Study of Chemical Vapor Infiltration Processing of Ceramic Composites," *Science*, **260** [5109] 789-92 (1993).
26. A. Morales-Rodriguez, P. Reynaud, G. Fantozzi, J. Adrien, and E. Maire, "Porosity Analysis of Long-Fiber-Reinforced Ceramic Matrix Composites Using X-Ray Tomography," *Scr. Mater.*, **60** [6] 388-90 (2009).
27. C. Chateau, L. Gelebart, M. Bornert, J. Crepin, E. Boller, C. Sauder, and W. Ludwig, "In Situ X-Ray Microtomography Characterization of Damage in SiC_f/SiC Minicomposites," *Compos. Sci. Technol.*, **71** 916-24 (2011).
28. H. A. Bale, A. Haboub, A. A. MacDowell, J. R. Nasiatka, D. Y. Parkinson, B. N. Cox, D. B. Marshall, and R. O. Ritchie, "Real-Time Quantitative Imaging of Failure Events in Materials Under Load at Temperatures Above 1,600°C," *Nat. Mater.*, **12** [1] 40-46 (2013).
29. <http://rsbweb.nih.gov/ij/>.
30. D. Kim, J. Lee, W. -J. Kim, and J. Y. Park, "Effect of Filament Winding Methods on Surface Roughness and Fiber Volume Fraction of SiC_f/SiC Composite Tubes," *J. Kor. Ceram. Soc.*, **50** [6] 359-63 (2013).